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High-Resolution Photoassociation Spectroscopy of Ultracold Ytterbium Atoms by Using the Intercombination Transition

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We observed high-resolution photoassociation spectra of laser-cooled ytterbium (Yb) atoms in the spin-forbidden $^1S_0 - ^3P_1$ intercombination line. The rovibrational levels in the 0_u^+ state were measured for red detunings of the photoassociation laser ranging from 2.9 MHz to 1.97 GHz with respect to the atomic resonance. The rotational splitting of the vibrational levels near the dissociation limit were fully resolved due to the sub-MHz linewidth of the spectra in contrast to previous measurements using the spin-allowed singlet transition. In addition, from a comparison between the spectra of ^{174}Yb and those of ^{176}Yb , a d -wave shape resonance for ^{174}Yb is strongly suggested.

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Photoassociation (PA) spectroscopy in laser-cooled atoms is a powerful technique for the accurate determination of the interatomic interaction including the scattering length, and has been successfully demonstrated for alkali atoms [1–3]. Recently PA spectroscopy has been reported for metastable He [4,5], alkaline-earth atoms Ca [6], Sr [7,8], as well as Yb [9], which is a rare-earth element with electronic structure similar to the alkaline-earth atoms. An alkaline-earth-like atom has a simple energy structure with the lack of the hyperfine structure in bosonic isotopes. The simple potential makes it easy to understand the interatomic interaction from PA spectra [10].

In all reports of the PA experiments so far, the dipole-allowed transition was used; e.g., $^2S_{1/2} - ^2P_{1/2,3/2}$ transitions for alkali atoms, $^1S_0 - ^1P_1$ transition for alkaline-earth-like atoms, and $^3S_1 - ^3P_{0,2}$ transitions for metastable He. For alkaline-earth-like atoms, the spin-forbidden intercombination $^1S_0 - ^3P_1$ transition is also useful for PA spectroscopy [11]. The narrow linewidth in the $^1S_0 - ^3P_1$ transition, in comparison with the broad one in the $^1S_0 - ^1P_1$ transition, results in high-resolution PA spectra. Furthermore, the optical tuning of the scattering length, the so-called optical Feshbach resonance, in the intercombination transition is expected to be achieved without excessive atom loss [12,13] in comparison with that in a strong allowed transition [14]. In order to successfully perform the PA experiment using the intercombination transition, however, we need to overcome the difficulties caused by the fact that a transition probability of a forbidden transition is several orders of magnitude smaller than that of an allowed one in general and that the homogeneous linewidth is much smaller than the Doppler width even at the low temperature of 100 μK in comparison with other PA experiments in spin-allowed transitions. Therefore, an experimental condition of high-density and ultralow temperature should be needed.

In this work we have successfully observed high-resolution PA spectra, typically sub-MHz, for the $^1S_0 - ^3P_1$ transition using the ultracold Yb atoms. The PA spectra are related with the free ($^1S_0 + ^1S_0$)-bound (0_u^+) transition shown in Fig. 1, where the 0_u^+ state connects to the $^1S_0 + ^3P_1$ asymptote in the dissociation limit. The long-range potential $U(R)$ in the 0_u^+ state of Yb_2 at interatomic distance R is determined by the resonant dipole-dipole interaction and the rotational term [6,7,11,15]

$$U(R) = D_e - \frac{C_3}{R^3} + \frac{\hbar^2[J(J+1) + 2]}{2\mu R^2}, \quad C_3 = \frac{3\hbar\lambda^3}{16\pi^3\tau}, \quad (1)$$

where D_e is the dissociation energy, J is the total angular momentum quantum number of the molecule, μ is the reduced mass of the molecule, λ is the wavelength of the $^1S_0 - ^3P_1$ transition, and τ is the radiative lifetime of the 3P_1 state. The rotational term of Eq. (1) is specific to the 0_u^+ state of the $^1S_0 + ^3P_1$ asymptote [11]. The narrow line-

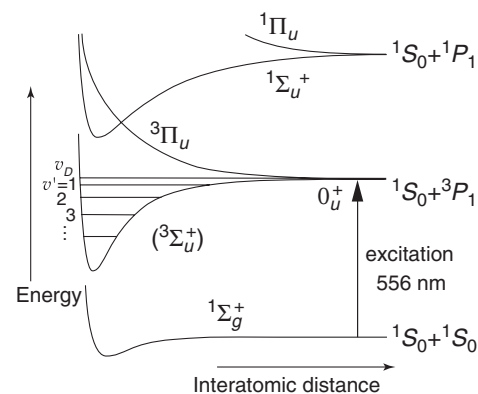


FIG. 1. Pair potential of Yb_2 . The PA light of 556 nm excites the two free 1S_0 atoms to the molecular excited level 0_u^+ which connects asymptotically to the $^1S_0 + ^3P_1$ state in the dissociation limit.

width of the spectra resulted in well-resolved rotational levels even near the dissociation limit. From the behaviors of the PA signals with $J = 1$ and $J = 3$ at various temperatures, we could obtain useful information about the ground-state scattering properties of Yb atoms.

The experimental setup and procedure are based on a magneto-optical trap (MOT) with the intercombination transition and a crossed far-off resonant trap (FORT) system [9,16,17]. The Zeeman-slowed Yb atoms with the strong dipole-allowed transition ($^1S_0 - ^1P_1$, 399 nm) were cooled and trapped in the MOT using the intercombination transition ($^1S_0 - ^3P_1$, 556 nm). The number of the atoms was about 10^7 at the temperature of about 40 μ K. The atoms were transferred into the crossed-FORT generated from cw diode-pumped solid-state laser systems whose wavelengths were 532 nm and beam waists of horizontal and vertical FORT were 14 and 11 μ m, respectively. About 10^6 atoms at the temperature of 180 μ K were trapped in the FORT. The initial potential depths of horizontal and vertical FORT beams were 630 μ K (corresponding to the FORT power of 5.0 W) and 53 μ K (0.2 W), respectively. Evaporative cooling was carried out by gradually decreasing the potential depth of the horizontal FORT beam to several μ K in about 6 sec. Typically, 10^4 atoms finally remained in the trap. The temperature was controlled from 2 to 30 μ K. After the evaporative cooling in the crossed FORT, the PA light focused to a 100 μ m diameter was applied for about 100 ms with the PA power controlled so that at least half of the trapped atoms remained. The PA light was obtained by dividing the laser light for the MOT and the frequency was controlled by acousto-optic modulators. The laser linewidth was much narrower than 100 kHz by frequency stabilization to an ultralow expansion cavity. The linewidth was small enough compared with the molecular linewidth of 364 kHz for the $^1S_0 + ^3P_1$ state. We observed the PA signals by measuring the number of the survived atoms with the absorption imaging method using the strong $^1S_0 - ^1P_1$ transition.

Figure 2 shows the obtained PA spectra of ^{174}Yb atoms. We observed 13 PA spectra of vibrational levels in the 0_u^+ state up to about 2 GHz red-detuning from the atomic resonance with the PA light intensity in the range of 6.5 $\mu\text{W}/\text{cm}^2$ to 69 mW/cm^2 at the temperature of typically 4 μ K. At this temperature only the s -wave collision contributes to the PA spectra. Therefore, the PA only occurs between the excited state with the rotational quantum number $J = 1$ and the ground state with $J_g = 0$ by the selection rule $\Delta J = \pm 1$. The vibrational quantum number from the dissociation limit was successfully assigned in the range of $v' = 7$ to 19, where $v' = v_D - v + 1 - (v_D \text{ modulo } 1)$, v is the standard vibrational quantum number, and v_D is the noninteger number corresponding to a hypothetical level at the dissociation limit (see Fig. 1) [6]. A novel feature of the observed PA spectra is the narrow linewidth, typically less than 1 MHz; e.g., 828 kHz for $v' = 9$ and 787 kHz for $v' = 19$ by fitting the theoretical

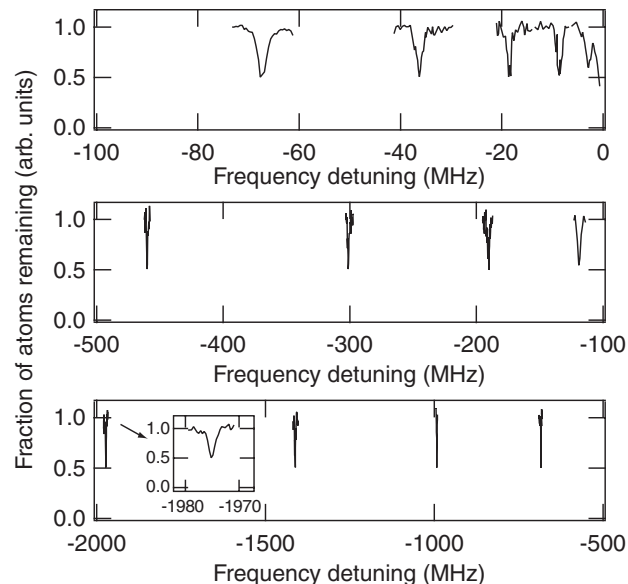


FIG. 2. PA spectra of ^{174}Yb atoms at about 4 μ K. The fraction of atoms remaining (solid lines) is shown against the detuning from the $^1S_0 + ^3P_1$ asymptote. The assigned v' is in the range of 7–19. An enlarged view for $v' = 19$ is shown in the inset.

spectral profile which consists of the Lorentz function and the energy distribution function [3,11]. The profile is slightly asymmetric at 4 μ K. The broadening effect observed by our previous PA work for the $^1S_0 - ^1P_1$ transition [9], presumably due to the predissociation effect, was not observed in the region of these detunings.

Figure 3(a) shows the observed resonance frequencies $\Delta(v)$ relative to the $^1S_0 + ^3P_1$ asymptote as a function of

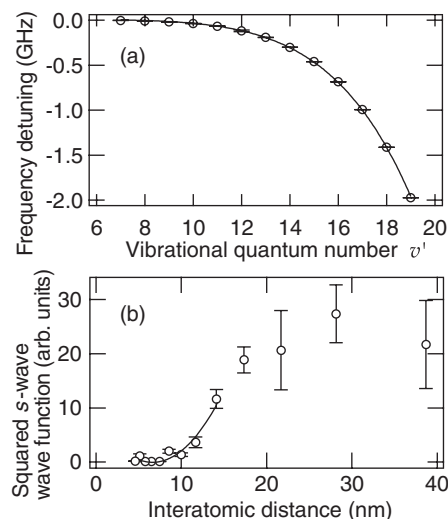


FIG. 3. (a) Resonance frequencies of the consecutive series as a function of the assigned vibrational quantum number v' in the range of 7–19 (open circles). Solid line is the fitted curve of the semiclassical formula of Eq. (2) in the text. (b) Open circles are the reconstructed squared wave function of the ground state for the s wave from the observed PA spectra, and the solid line is fitted by a quadratic function.

the assigned numbers v' in the range of 7 to 19. The vibrational energy structure $E(v)$ near the dissociation limit can be given by the semiclassical formula [18]

$$E(v) = D_e - X_0(v_D - v)^6 + X'_0(v_D - v)^4[J(J+1) + 2],$$

$$X_0 = \frac{h^6}{\mu^3 C_3^2} \left[\frac{\Gamma(4/3)}{2\sqrt{2}\pi\Gamma(5/6)} \right]^6, \quad X'_0 = \frac{\bar{X}_0}{\mu^3 C_3^2}, \quad (2)$$

where \bar{X}_0 is the numerical factor given in Ref. [18]. The curve fitted with the function $\Delta(v) = [D - E(v)]/h$ is also shown in Fig. 3(a), where only v_D is the fitting parameter. We note that the experimental data are well fitted by the curve with the lifetime $\tau = 874$ ns given in Ref. [19] for the intercombination transition.

The signal intensities of the observed PA resonances at sufficiently low temperatures offer information of the squared s -wave wave function in the ground-state scattering on the basis of the reflection approximation [3,8,20]. The reflection approximation can be well applied also for the Yb atoms in the $^1S_0 - ^3P_1$ transition since the interatomic potential of the excited state is quite different from that of the ground state, in contrast to the case of Ca atoms where both the excited and the ground-state potentials are expressed as $-C_6/R^6$ [11]. The PA rate is proportional to $I_{PA} n_0 F(\Delta)$ in the perturbative low intensity regime [3], where the free-bound Frank-Condon factor $F(\Delta)$ is given under the reflection approximation by

$$F(\Delta) = \frac{\partial E_v}{\partial v} \frac{1}{d_C} |\Psi_g(R_C)|^2, \quad (3)$$

where I_{PA} is the PA light intensity, n_0 is the atom density, $\Psi_g(R)$ is the wave function of the ground state, Δ is the frequency detuning from the dissociation limit, R_C is the Condon point, and $d_C = |\frac{d}{dR}[V_e(R) - V_g(R)]|_{R=R_C}$ is the difference of slopes between the excited state $V_e(R)$ and the ground state $V_g(R)$ at R_C . Figure 3(b) shows the squared wave function with the energy distribution from the PA spectra using Eq. (3). To deduce the squared wave function in Fig. 3(b), we have used the data in the perturbative regime. The squared wave function around the node can be fitted by a quadratic function of $(R - R_0)^2$, which is shown in Fig. 3(b). From the fitting curve we derived the last node $R_0 = 6.7$ nm [21].

We also observed the PA spectra at a higher temperature. Figs. 4(a) and 4(b) show the PA spectra at 25 μ K with PA light intensities of 0.2 and 90 mW/cm², respectively. The assigned vibrational quantum numbers in Figs. 4(a) and 4(b) are $v' = 10$ ($R_C = 17.3$ nm) and $v' = 16$ ($R_C = 6.51$ nm), respectively. These data clearly show well-resolved two rotational resonances corresponding to $J = 1$ and $J = 3$. The separation between the rotational levels in each vibrational state is consistent with theoretical formula of Eq. (2) [18]. The higher-order rotational resonances such as $J = 5$ were not found around expected frequency detunings from Eq. (2). The solid lines are fitted by the theoretical spectral profile. The rotational spectra of

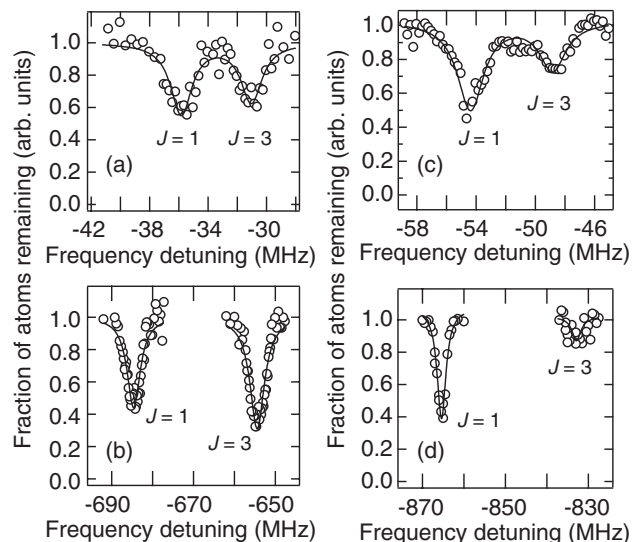


FIG. 4. Rovibrational PA spectra at about 25 μ K; (a),(b) ^{174}Yb and (c),(d) ^{176}Yb . Open circles are the fraction of the atoms remaining. Solid lines are fitted curves of the theoretical profile. The assigned vibrational quantum numbers are (a),(c) $v' = 10$ and (b),(d) $v' = 16$.

$J = 3$ comes from the atoms in the d -wave scattering state ($J_g = 2$).

The potential curve for the d -wave scattering in the long range is determined by the centrifugal barrier and the van der Waals interaction of $-C_6/R^6$. Figure 5(a) shows the s - and d - partial-wave potential of Yb in the ground state by assuming $C_6 = 1000$ a.u. [22]. The calculated height of the d -wave potential barrier is about 300 μ K which is much higher than the kinetic energy of the atoms in the experiment, and so it is natural to expect that the atoms in d -wave scattering cannot come closer than around 6.0 nm due to the centrifugal barrier. In spite of the high

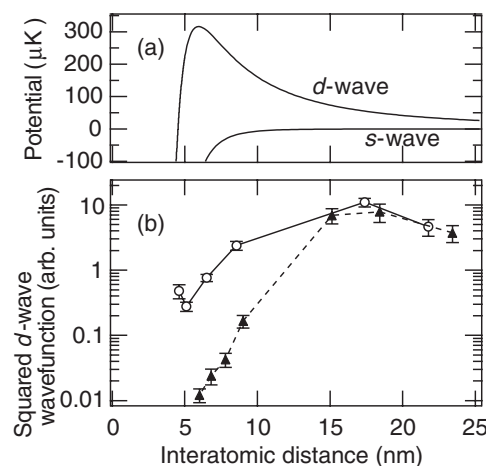


FIG. 5. (a) Partial-wave potential as a function of interatomic distance with $C_6 = 1000$ a.u. (b) The reconstructed squared wave functions of the ground state for the d wave from the observed PA spectra for $J = 3$ of ^{174}Yb (open circles with solid line) and ^{176}Yb (closed triangles with dashed line).

potential barrier the PA spectra originated from d wave were observed as shown in Fig. 4(b). The PA rate of $J = 3$ is rather larger than that of $J = 1$.

In order to examine the behavior of the d wave we also performed the PA spectroscopy for ^{176}Yb . The present method can be easily applied to other bosonic isotopes without essential modification of experimental parameters [23]. Figs. 4(c) and 4(d) show PA spectra of ^{176}Yb atoms at 25 μK with PA intensities of 0.3 and 900 mW/cm^2 , respectively; (c) $\nu' = 10$ ($R_C = 15.1$ nm) and (d) $\nu' = 16$ ($R_C = 6.02$ nm). The frequency separation between $J = 1$ and $J = 3$ is also consistent with Eq. (2). The higher-order rotational resonances were not found as well. Striking difference between PA signal for $J = 3$ of ^{174}Yb and those of ^{176}Yb was observed at $R_C \leq 10$ nm. In the region near $R_C = 6$ nm the PA rate for ^{174}Yb was 2 orders of magnitude larger than that for ^{176}Yb . Figure 5(b) shows the squared ground-state wave functions for the d wave of ^{174}Yb and ^{176}Yb .

We note that the validity of the reflection approximation is independent of the order of the partial wave [24], so we can apply the expression of Eq. (3) similarly for the d wave. Thus reconstructed d -wave wave function of ^{176}Yb is consistent with the conventional exponential attenuation in the potential barrier. On the other hand, the attenuation of the wave function of ^{174}Yb is much weaker. The magnitude of the squared d -wave wave function of ^{174}Yb is about 2 orders of magnitude larger than that of ^{176}Yb . The behavior can be explained by an effect of the d -wave shape resonance in the ground-state potential for ^{174}Yb [25,26]. In the region of the low temperature less than the centrifugal barrier, d -wave scattering should be reduced exponentially in atomic collision without any shape resonance. We note that the existence of the d -wave shape resonance of ^{174}Yb may play an important role in the efficient evaporative cooling of ^{174}Yb , which has been successfully evaporatively cooled to BEC [16].

In conclusion, we reported the first observation of PA spectra in the intercombination transition by using ultra-cold Yb atoms. We observed high-resolution PA spectra with sub-MHz linewidths in rovibrational states even in the long-range interatomic distances. The d -wave shape resonance for ^{174}Yb is strongly suggested in comparison with ^{176}Yb .

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